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(54) Title: USE OF CATALYSTS		
(57) Abstract		
The present invention relates to the use of a catalyst in a catalytic combustion, which catalyst is produced by the coating of a carrier with a suspension of noble metal particles in a microemulsion. The carrier is distributed on a monolith. The invention also relates to a catalyst comprising an active phase which by microemulsion technique has been deposited on a carrier. The active phase comprises noble metal particles. The carrier comprises a metal oxide and is distributed on a monolith. The invention also relates to a method of producing a catalyst as defined above by microemulsion technique, wherein bimetallic particles are produced by the blending of not reduced noble metal particles in microemulsion and then the reduction thereof.		

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USE OF CATALYSTSField of invention

The present invention relates to the use of a particular class of catalysts in combustions as well as to new catalysts belonging to said class. The invention also  
5 relates to a method of producing said catalysts.

Background of the invention

The interest in alternative fuels began in the early 1970's with the so called energy crisis. The increased  
10 awareness of the environment during the past decades has focussed the interest on the search for alternative fuels, which are harmless to the environment. The combustion of these fuels has to be possible to perform with a high performance as well as a good selectivity to provide  
15 as low as possible a level of poisonous or unhealthy substances in the effluents.

Catalytic combustion increases the activity of the process of combustion at the same time as the selectivity may be improved, whereby the amount of many ecologically  
20 harmful substances in the effluents is reduced, or even eliminated.

In Sweden, it has been found that an alternative fuel for engine running, which is harmless to the environment as well as of economical interest, is alcohol based fuels, such as ethanol. This choice depends on  
25 the access to raw material in the country as well as the resources to produce ethanol. A catalytic combustion of the products of ethanolic combustion is therefore a for the future very promising process, since it together with  
30 the appropriate catalyst should be possible to design as an extraordinarily active and selective process, which thereby is harmless to the environment.

Ethanol is already used as a fuel for buses driven on trial in Stockholm. These buses are equipped with

diesel engines. It is well known that ecologically harmful substances are formed when oil and petrol are combusted. Similarly, undesirable substances, such as acetaldehyde and acetic acid, are formed when ethanol is 5 combusted.

The effluents from petrol engines are these days taken care of by car effluent catalysts, which, inter alia, convert NO to N<sub>2</sub> and CO to CO<sub>2</sub>. These catalysts contain noble metals, such as Pt and Rh, deposited on a 10 carrier which often is composed of Al<sub>2</sub>O<sub>3</sub>.

The same class of catalysts are at the moment used for the purification of the effluents from diesel engines, wherein ethanol is used as a fuel. The effluents from ethanol vehicles contain non-combusted ethanol and 15 acetaldehyde, which in turn may be converted to acetic acid, which is irritating due to its particular odour.

To avoid that these substances are let out in effluents, efforts have been made to find suitable oxidation catalysts. Previously, mainly catalysts based on noble 20 metals have been used. However, base metal oxides have been examined as well.

Another area where catalytic combustion is of interest is combustion of fuels in such applications as industrial boilers, gas turbine burners and heat generating systems, i.e. systems that primarily aim at energy 25 recovery. However, the requirements within this field on the properties of the material and complex chemical reaction systems make this one of the most difficult applications of catalytical combustion.

30

#### Technical problem

When ethanol is used as a fuel in diesel engines it has been shown, that the catalysts that exist today work satisfactorily during the driving. However, problems 35 arise in situations when the engine is running idle, for instance at bus stops or when in a traffic jam. The

reason is that in such situations, the temperature of the catalyst decreases, whereby the activity as well as the selectivity are deteriorated substantially. Accordingly, acetic acid and acetaldehyde, as well as some non-combusted ethanol, exist in the effluents discharged in the city environment.

Thus, a substantial need of a more specific catalyst, which inter alia has the capacity of working efficiently at low temperatures, exists, since the temperature of the effluents from an engine running idle is 250°C or less.

To specifically design a catalyst which in an efficient way is able to take care of the effluents formed in ethanolic combustion it is necessary to identify the combustion products to work out which the products formed at low temperatures are. Thereafter, it is possible to develop the capacities of the catalyst to convert these products to effluents which are less harmful to the environment. As mentioned above, effluents from ethanol vehicles contain non-combusted ethanol and acetaldehyde and these substances, together with the acetic acid, have to be removed. Hitherto, the efforts to develop catalysts which enables this efficiently at low temperatures have failed.

It has been shown that in the use of catalysts for combustion for the purpose of energy recovery a pre-catalyst, which ignites at a low temperature, is of great importance.

30 Description of the invention

The present invention essentially solves the above defined problem by providing the use of a new class of catalysts for catalytic combustions. Said catalysts are produced by the coating of a carrier with a suspension of noble metal particles in a microemulsion, the carrier further being distributed on a monolith. The invention

further relates to said catalysts and to a method for the production thereof.

Some catalysts of this kind are known per se from EP 81900804.6. However, the present use is not suggested, 5 and even less the extraordinarily results accomplished by the catalyst according to the invention in said use.

Detailed description of the invention

More specifically, the invention relates to a new 10 use of a catalyst in combustions, wherein the catalyst has been produced by the coating of a carrier with a suspension of noble metal particles in a microemulsion. The carrier has been distributed on a monolith as a support and in order to maximize the contact area between the 15 active or catalytic parts of the catalyst on the carrier and the combustion gases.

In one embodiment of the invention a carrier, which has been coated with a monolayer of the noble metal particles, is used. Thereby the contact area relative to the 20 effluents is increased substantially.

In another embodiment of the invention a catalyst as mentioned above is used, wherein the noble metal particles are monolithic, i.e. each particle is composed of one metal species. Also included are embodiments, where the 25 catalyst includes several metal species, as long as each particle does not include more than one species.

In an alternative embodiment of the invention the noble metal particles are bimetallic, i.e. composed of two metals.

30 In another embodiment of the invention a catalyst is used, wherein the carrier has been coated with noble metal particles from the Pt group, preferably Pt and/or Pd.

In a certain embodiment of the invention said noble metal particles are Pt and Pd, which have been coreduced 35 in the microemulsion. By the simultaneous reduction of the metals, blended together, surprisingly, an activity is achieved, which is substantially higher than that

achieved when the metals are first reduced each one separately and thereafter blended.

In one embodiment of the invention the carrier is a monolith, such as  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  or  $\text{SiC}$ , preferably  $\text{Al}_2\text{O}_3$ .

5 In another embodiment of the invention a monolith is used, which is composed of a metal, a metal alloy or inorganic oxides. The geometrical shape thereof is such, that combustion gases, or effluents, can pass without any substantial pressure drop. A preferred monolith is produced from cordierite ( $2\text{MgO} \cdot 5\text{SiO}_2 \cdot 2\text{Al}_2\text{O}_3$ ). A preferred shape 10 is a cylinder with a large amount of square channels for the reduction of said pressure drop to a minimum.

In one embodiment of the invention the catalytic combustion is the combustion of alcohol based fuels, especially ethanol. The use of the catalyst according to 15 the invention, whose activity is surprisingly substantially higher than the previously known catalysts, especially at low temperatures, has been proven particularly advantageous in those diesel engines, with which the ethanol fueled city buses are equipped at present.

In another embodiment of the invention the catalytic combustion is combustion in gas turbines, industrial boilers or heat generating systems. The low flash point in the use of the catalyst according to the invention should 25 under these conditions be especially advantageous.

Another aspect of the invention is a catalyst, which comprises an active phase, which has been deposited on a carrier by microemulsion technique. The active phase comprises noble metal particles and the carrier comprises 30 a metal oxide. Also in this case the carrier has been distributed on a monolith. The catalyst according to the invention has proven to be especially suitable for combustion reactions, especially of the kind that has been discussed above in context with the use according to the 35 invention, such as the combustion of alcohol based or other organic materials.

In one embodiment of this aspect of the invention the carrier has been coated with a monolayer of the noble metal particles.

5 In another embodiment of this aspect the noble metal particles are monometallic.

In an alternative to this embodiment the noble metal particles are bimetallic.

10 In a particular embodiment the catalyst according to the invention is composed of noble metal particles, which for instance belong to the Pt group, preferably Pt and/or Pd. In one particular embodiment the noble metal particles are Pt and Pd, which have been coreduced in the microemulsion, i.e. they have been admixed before the reduction thereof was performed.

15 In another embodiment the metal oxide of the carrier is Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> or SiC, Al<sub>2</sub>O<sub>3</sub> being preferred at present thanks to its advantageous ageing properties.

20 In a special embodiment of the catalyst according to the invention the monolith is a metal, a metal alloy or inorganic oxides.

25 According to a further aspect the invention relates to a method of producing such a catalyst as is described above. The catalyst is manufactured by a microemulsion technique which is known per se. However, according to the invention bimetallic particles are produced by the blending of non-reduced noble metal particles in the microemulsion and then the reduction thereof.

30 According to a special embodiment of the method according to the invention the noble metal particles belong to the Pt group, preferably Pt and Pd.

#### EXAMPLE

##### Production of the catalyst

35 Three microemulsions were produced for the experiments, one Pt microemulsion and one Pd microemulsion as well as one microemulsion which contained both Pt and Pd. From a monolith, with 142,4 g/dm<sup>3</sup> of Al<sub>2</sub>O<sub>3</sub> carrier, test

specimens were cut out. The sizes of the test specimens were designed as to fit in an existing test plant. The monolith had a cell density of 62 cells/cm<sup>2</sup>.

5 The metal salts were reduced and deposited on the test specimens. The volume of the monolith specimens are estimated to 11000 mm<sup>3</sup>, or 11x10<sup>-3</sup> dm<sup>3</sup>.

10 The monoliths were produced with an metal charge according to Table 1. These metal levels were chosen to enable a relevant comparison with already performed tests of catalytic activity.

The term "Pt-Pd" denotes that the metal salts were coreduced, i.e. both salts were present at the reduction. "Pt+Pd" denotes that each metal salt was reduced in its own microemulsion and that they were then blended.

15

Table 1

Catalyst	Metal concentration on monolith, g/dm <sup>3</sup>	Metal concentration on monolith, mmol/dm <sup>3</sup>
Pt	1,8	9,5
Pd	1,0	9,5
Pt, Pt	0,9, 0,5 (1,4 in total)	4,8, 4,8 (9,5 in total)

20 The carrier consists of  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> (Condea PX140) with a specific area of about 140 m<sup>2</sup>/g. These values are valid for catalysts produced by impregnating technique as well as catalysts produced by microemulsion technique.

25 Catalytic reaction  
The experimental design enables catalytic tests of gas mixtures, whose composition is such that it resembles the effluents produced in ethanol combustion in a diesel engine.

Table 2

Substance	Concentration
O <sub>2</sub>	10 vol%
H <sub>2</sub> O	10 vol%
CO <sub>2</sub>	6,5 vol%
N <sub>2</sub>	Balance
CO	300 ppm
NO	600 ppm
C <sub>2</sub> H <sub>5</sub> OH	200 ppm

- 5 The catalyst is tested at an excess of air of 100% ( $\lambda=2$ ) in a gas mixture, whose composition is given in Table 2. The experiments are performed at between 75°C and 500°C with a space velocity of 100000h<sup>-1</sup>. All of the catalysts were pretreated. The pretreatment implies that the catalyst is heated from room temperature to 500°C, with a heating rate of 100/minute. During the heating, the reaction mixture is passed over the catalyst in the reactor. The catalyst is then cooled with a gas mixture, the composition of which is 10% of O<sub>2</sub>, 10% of H<sub>2</sub>O and 80% of N<sub>2</sub>.
- 10 15 During cooling, the space velocity of the gas mixture is 25000 h<sup>-1</sup>.

Measuring equipment

- The concentration of the total amount of hydrocarbon, nitrogen oxide and carbon monoxide in the discharge was analyzed on line. To this end, conventional instruments for analysis were used. The total amount of hydrocarbon was analyzed with a flame ion detector (FID). Nitrogen oxide was analyzed on a chemiluminescence detector (CLD) and carbon dioxide was measured with a non-dispersive infra red instrument (NDIR). A detailed

analysis of oxygen, nitrogen, carbon dioxide, oxidizing substances and hydrocarbons was performed on line in a semicontinuous manner. This was achieved on a gas chromatograph equipped with thermic conductivity, flame ionization and far ultra violet detectors.

### Results

In Table 3, the catalytic experiments are summarized. Values commonly used for the characterization of catalysts have been tabulated. The light-off temperature presented refers to the temperature when a conversion of 50% of the specific component exists.

Table 3

15

Catalyst	Metal	Light-off temperature for ethanol (50% conv.), °C	Light-off temperature for CO (50% conv.), °C	Max conv. of ethanol to acetaldehyde, %	Max conv. of ethanol to acetic acid, %
Impregnated	Pt	153 (149 <sup>1</sup> )	167	30	2
	Pd	213 (203 <sup>1</sup> )	190	33	0,40
	Pt, Pd	130 <sup>1</sup>	(135 <sup>1</sup> )	-	not det.
Micro-emulsion	Pt	116	166	35-45	not det.
	Pd	164	176	55	not det.
	Pt-Pd	111	152	45	not det.
	Pt+Pd	164	173	45	not det.

<sup>1</sup> Results from catalysts deposited on TiO<sub>2</sub> and produced by impregnation technique.

The following reaction steps occur in the oxidation of ethanol:



Thus, acetic acid or acetaldehyde is formed in the oxidation of ethanol. It is evident that acetaldehyde is formed during the course of the reaction with all catalysts. On the contrary, no detectable amount of acetic acid is formed. No acetic acid could be detected when the catalysts were produced by microemulsion technique either.

### Catalytic oxidation of ethanol

The catalysts produced by microemulsion technique which are the most active ones are the catalysts containing Pt (i.e. Pt, Pt-Pd, Pt+Pd). The light-off temperatures for these catalysts are measurably lower than for the monolith which was coated with Pd only. It is true that catalysts produced by impregnation technique exhibit the same interrelated differences depending on the metal compositions, but the activities thereof are generally lower than those of the corresponding produced by microemulsion technique.

The light-off temperatures for microemulsion produced catalysts are generally lower than for the corresponding catalysts produced by impregnation.

35 For microemulsion produced Pt, Pd and Pt-Pd

catalysts the light-off temperatures are respectively 37, 39 and 20 °C lower than for the corresponding impregnated catalysts (Table 3).

When comparing impregnated catalysts with TiO<sub>2</sub> as a carrier with the corresponding γ-Al<sub>2</sub>O<sub>3</sub> catalysts, it is obvious that an impregnated bimetallic Pt-Pd catalyst with a TiO<sub>2</sub> carrier yields a 25°C lower light-off temperature than a γ-Al<sub>2</sub>O<sub>3</sub> catalyst does. Microemulsion produced Pt catalysts on Al<sub>2</sub>O<sub>3</sub> still yield a light-off temperature, which is 37°C lower than that for the impregnated Pt catalysts on TiO<sub>2</sub>.

From these results, it is obvious that microemulsion produced catalysts are active at lower temperatures than the corresponding impregnated catalysts, irrespective of the metal composition. Pt and Pt-Pd are the most active ones. When Al<sub>2</sub>O<sub>3</sub> is replaced with TiO<sub>2</sub> in the impregnated catalysts, a light-off temperature, which is 5°C lower than with the Pt-Pd catalysts on Al<sub>2</sub>O<sub>3</sub> produced by microemulsion technique, results.

20

#### Formation of acetaldehyde

It is observed that for all of the above discussed catalysts acetaldehyde is formed during the course of the reaction, unregarded which method of production of the catalyst is used. Catalysts produced in a microemulsion convert a larger amount of ethanol into acetaldehyde than impregnated catalysts do. The yield is 10-15% higher than with impregnated catalysts, unregarded the metal composition. The Pd catalyst provides the highest yield of 55%.

30

In comparing the temperatures at which acetaldehyde begins to decrease, it is observed that for microemulsion produced Pt catalysts, the acetaldehyde conversion decreases about 35°C earlier than the corresponding impregnated catalyst.

35

Formation of acetic acid

No acetic acid could be detected with microemulsion produced catalysts. With impregnated catalysts acetic acid is formed with a yield of 2% with the Pt catalyst 5 and 0,38 with Pd. The detection limit for acetic acid with the present instrumentation is about 0,5 ppm.

Yield of CO

Of the microemulsion produced catalysts the Pt-Pd 10 catalyst had the lowest light-off temperature, also when it comes to the oxidation of CO. The light-off temperature for this catalyst is 14°C lower than the best value presented in connection with the impregnated catalysts. On comparison of the Pd catalysts it appears that the 15 microemulsion produced Pd catalyst has a light-off temperature which is 14°C lower than that of the corresponding impregnated Pd catalyst. The light-off temperatures for the Pt catalysts are approximately equivalent for the two classes.

20

Discussion

The studies of the ethanol oxidation on different catalysts show that a number of different factors have influence on the activity of the catalyst. These factors 25 are *inter alia* the method of production, the nature of the metal and the metal composition. The carrier material also has an influence on the activity of the catalyst.

As regards the nature of the metal it appears that the method of production does not influence upon the difference between the Pt and Pd catalysts, i.e. that Pt is more active than Pd in ethanol oxidation. This might be explained by the fact that Pd forms PdO at a temperature lower than that at which Pt forms PtO. The respective metal oxide is assumed to be less active than Pt<sup>0</sup> and Pd<sup>0</sup>.

35 The catalyst of Pt+Pd, which was produced by the blending together of microemulsion produced Pt and Pd particles, did not exhibit the same activity as the Pt-Pd

catalyst. The Pt-Pd catalyst was produced by reduction in a microemulsion which contained metal salt of both Pt and Pd. The catalytic activity of the Pt+Pd catalyst is in the same range as the corresponding catalyst with Pd particles only, i.e. it has a high light-off temperature as compared to the Pt catalyst. On the contrary, the Pt-Pd catalyst has a light-off temperature which is comparable to that of the corresponding Pt catalyst. This implies that to obtain a high activity with the bimetallic catalysts, the metal particles have to be produced so that the metals are alloyed or blended together.

It is also recognized that the bimetallic catalysts of Pt and Pd lower the light-off temperature during CO oxidation, which may be of interest within other applications, where catalysts are used, i.e. three way catalysts.

#### Conclusion

The catalysts produced in a microemulsion have been shown to exhibit higher activities in ethanol oxidation than those, which have been produced by impregnation technique. In these studies, only the activities of catalysts produced by  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> as carriers have been compared. However, studies of catalysts produced by impregnation technique show that TiO<sub>2</sub> may be of interest as an alternative to  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.

CLAIMS

1. The use of a catalyst in catalytic combustion, which catalyst has been produced by the coating of a carrier with a suspension of noble metal particles in a microemulsion, the carrier being distributed on a monolith.
2. The use according to claim 1, wherein the carrier has been coated with a monolayer of the noble metal particles.
- 10 3. The use according to any one of claims 1 and 2, wherein the noble metal particles are monometallic.
4. The use according to any one of claims 1 and 2, wherein the noble metal particles are bimetallic.
- 15 5. The use according to any one of the preceding claims, wherein the carrier has been coated with particles of noble metal belonging to the Pt group, preferably Pt and/or Pd.
6. The use according to any one of claims 1-2 and 4-5, wherein the noble metal particles are Pt and Pd, which have been coreduced in the microemulsion.
- 20 7. The use according to any one of the preceding claims, wherein the carrier is a metal oxide, preferably  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  or  $\text{SiC}$ .
8. The use according to any one of the preceding 25 claims, wherein the monolith is from a metal, a metal alloy or inorganic oxides.
9. The use according to any one of the preceding claims, wherein the catalytic combustion is the combustion of alcohol based fuels, especially ethanol.
- 30 10. The use according to any one of claims 1-8, wherein the catalytic combustion is a combustion in gas turbines, industrial boilers or heat generating systems.
11. A catalyst, especially for combustion reactions, comprising an active phase, which has been deposited on a 35 carrier by microemulsion technique, the active phase comprising noble metal particles, the carrier comprising

a metal oxide and the carrier having been distributed on a monolith.

12. A catalyst according to claim 11, wherein the carrier has been coated with a monolayer of the noble  
5 metal particles.

13. A catalyst according to any one of claims 11 and 12, wherein the noble metal particles are monometallic.

14. A catalyst according to any one of claims 11 and 12, wherein the noble metal particles are bimetallic.  
10

15. A catalyst according to any one of the preceding claims, wherein the particles are of a noble metal belonging to the Pt group, preferably Pt and/or Pd.  
15

16. A catalyst according to any one of claims 11-12 and 14-15, wherein the noble metal particles are Pt and Pd, which have been coreduced in the microemulsion.  
15

17. A catalyst according to any one of the preceding claims, wherein the metal oxide in the carrier is  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  or SiC.

18. A catalyst according to any one of the preceding  
20 claims, wherein the monolith is from a metal, a metal alloy or inorganic oxides.

19. A method of producing a catalyst according to any one of the preceding claims by microemulsion technique, wherein bimetallic particles are produced by  
25 blending non-reduced noble metal particles in a micro-emulsion and then reducing the same.

20. A method according to claim 19, wherein the bi-metallic particles are of noble metals belonging to the Pt group, preferably Pt and Pd.

## AMENDED CLAIMS

[received by the International Bureau on 28 January 1997 (28.01.97);  
original claims 1-20 replaced by claims 1-20 (2 pages)]

1. The use of a catalyst in catalytic combustion of alcohol based fuels, which catalyst has been produced by  
5 the coating of a carrier with a suspension of noble metal particles composed of two metals in a microemulsion, the carrier being distributed on a monolith.
2. The use according to claim 1, wherein the carrier has been coated with a monolayer of the noble metal  
10 particles.
3. The use according to any one of the preceding claims, wherein the carrier has been coated with particles of noble metal belonging to the Pt group, preferably Pt and/or Pd.
- 15 4. The use according to any one of the preceding claims, wherein the noble metal particles have been coreduced in the microemulsion.
5. The use according to claim 4, wherein the noble metal particles are Pt and Pd.
- 20 6. The use according to any one of the preceding claims, wherein the carrier is from the group consisting of Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and SiC.
7. The use according to any one of the preceding claims, wherein the monolith is from a metal, a metal  
25 alloy or inorganic oxides.
8. The use according to any one of the preceding claims, wherein the catalytic combustion is the combustion of ethanol.
9. The use according to any one of claims 1-6,  
30 wherein the catalytic combustion is combustion in diesel engines, gas turbines, industrial boilers or heat generating systems.
10. A catalyst, especially for combustion reactions, comprising an active phase, which has been deposited on a  
35 carrier by microemulsion technique, the active phase comprising noble metal particles composed of two metals,

which have been coreduced in the microemulsion, the carrier having been distributed on a monolith.

11. A catalyst according to claim 10, wherein the carrier has been coated with a monolayer of the noble metal particles.

12. A catalyst according to any one of claims 10 and 11, wherein the particles are of a noble metal belonging to the Pt group, preferably Pt and/or Pd.

13. A catalyst according to any one of claims 10-12, wherein the carrier is from the group consisting of  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{SiC}$ .

14. A catalyst according to any one of claims 10-13, wherein the monolith is from a metal, a metal alloy or inorganic oxides.

15. A method of producing a catalyst according to any one of claims 10-14 by microemulsion technique, wherein particles composed of two metals are produced by blending non-reduced noble metal particles in a microemulsion and then reducing the same.

20 16. A method according to claim 15, wherein the particles composed of two metals are of noble metals belonging to the Pt group, preferably Pt and Pd.

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INTERNATIONAL SEARCH REPORT

International application No. PCT/SE 96/01070
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**A. CLASSIFICATION OF SUBJECT MATTER**

**IPC6: B01J 23/38, B01D 53/92, F01N 3/28**  
According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

**IPC6: B01J, B01D, F01N**

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

**SE,DK,FI,NO classes as above**

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4425261 A (PER STENIUS ET AL), 10 January 1984 (10.01.84), column 1, line 1 - line 28; column 6, line 7 - line 44 ---	1-20
Y	US 5179059 A (RAINER DOMSELE ET AL), 12 January 1993 (12.01.93), claim 24, abstract -----	1-20

Further documents are listed in the continuation of Box C.

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Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. +46 8 666 02 86	Authorized officer  Johan von Döbeln Telephone No. +46 8 782 25 00

**INTERNATIONAL SEARCH REPORT**  
Information on patent family members

28/10/96

		International application No.
		PCT/SE 96/01070

Patent document cited in search report	Publication date	Parent family member(s)		Publication date
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